



An experimental method for measuring the radon-222 emanation factor in rocks

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Abstract

An experimental method, based on a 21-day accumulation technique, is proposed for measuring the radon-222 emanation factor in undisturbed consolidated materials. The leakage rate is determined from the form of the radon growth curve in the measurement chamber. It was comparable to the radon decay. In order to obtain the “true” radon emanation factor, the thickness of the sample must be less than the radon diffusion length in the porous material. The method was used to measure the radon emanation factor in water-saturated claystones (argillites). The radon emanation factor, determined from experiments on a rock sample with a thickness of 5 mm, was 15%, a value typical for this kind of material.

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1. Introduction

Radon-222 has been widely measured in soil gas in the air of caves, tunnels and other underground cavities in order to detect zones of enhanced permeability induced by fractures or faults that affect geological formations (King et al., 1996; Gascoyne et al., 1993; Monnin and Seidel, 1997). For characterizing a potential site of underground waste storages, this easily detected radioactive gas could be used to help determining the confining properties of a geological formation, and, ultimately, its suitability as an underground repository.

Radon flux exhalation at the surface of a porous material results from two processes: The emanation, which corresponds to the release in the pore space of radon atoms formed by the radioactive decay of radium-226 atoms present in solid particles, and the migration by diffusion and advection in the porosity (pores, macro-pores, fractures). The radon emanation factor is defined as the ratio between the number

of radon atoms entering the free pore space and the total number of radon atoms formed (Tanner, 1980). In order to distinguish the contribution of a fracture to exhalation from the rock matrix source term, the radon emanation factor in the rock matrix must be determined.

Most emanation factor measurements are based on the accumulation technique. The sample material is placed in a closed container; some days after sealing, e.g. about 2 weeks for Moore and Moore (1984) and 1 month for Greeman and Rose (1996), a sample of gas is drawn into an evacuated scintillation cell to measure its radon activity. The radon emanation factor is thus deduced from the concentration reached in the air of the closed container at the end of the buildup period. This may, however, be affected by leakage from the sealed container and “back diffusion” into the sample material. In order to reduce these artefacts, Stranden (1983) proposed reducing the sampling time (< 20 h) and deducing the radon emanation factor in crushed samples from the slope of the radon concentration increase in the closed chamber. Because of the detection limit of the radon concentration measure, this method is not suitable for consolidated materials such as rocks, with low radium contents and low radon diffusion lengths.

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Baretto (1975) reported radon emanation factors measured with the accumulation method in crushed and sieved rock samples. This paper described an experimental method, which was specifically designed to measure the radon emanation factor in undisturbed rock samples or in other consolidated porous materials such as concrete.

The proposed method has the advantage of being based on a simple modification of an instrument that is commercially available. It is an accumulation technique, but in our case the radon increase in the closed chamber containing the sample is continuously monitored by an ionization chamber without disturbance induced by gas sampling. The first part describes the experimental method. Results of experiments made using claystone (argillites) samples are described and discussed in the second part.

2. Description of the method

2.1. The experimental assembly

The container is a hollow metallic cylinder. During the measurement, the cylinder is screwed onto the radon diffusion entry window of a commercially available ionization chamber (the Genitron Instruments AlphaGUARD). A rubber gasket seals the two parts of the measurement chamber. The total void volume in the measurement chamber is 964 cm^3 , which includes the ionization chamber and cylindrical chamber volumes.

The rock sample consists of a cylindrical core, surrounded with a polymer resin layer (epoxy LANKO 723), impervious to radon, except for one face (Fig. 1). The diameter of the rock sample is 3.5 cm. The total diameter of the block sample is close to 5.7 cm. The thickness of the sample must be optimized to meet two conditions; it must be small compared with the radon diffusion length in the porous medium studied, and as large as possible to get a large source of radon, which reduces the uncertainty in the radon concentration measurement. Given the radon diffusion length in unweathered rock matrix, which is generally of the order of 1 cm (Tanner, 1980), metal cylinders are added inside

the cylindrical chamber in order to minimize the total void volume in the measurement chamber, thus improving the sensitivity of the method. This volume is limited by the volume of the ionization chamber (560 cm^3), which cannot be reduced. The experiment assembly is illustrated in Fig. 1

2.2. Determination of the radon emanation factor

The radon concentration in the measurement chamber is monitored every hour until the steady state is reached. Because of its short half-life, the contribution of radon-220 to the total radon concentration measured in the ionization chamber is negligible, unless the thorium concentration in the sample is two orders of magnitude higher than its radium content. In this case, a void volume between the sample and the entry window of the ionization chamber must be maintained. Hereafter, the term radon designs only the isotope 222.

Based on work of Samuelson and Pettersson (1984), we assume that the exhalation rate from the sample is constant during the accumulation period. If both the leakage in the measurement chamber and the radon concentration in air outside are fairly constant, then the radon concentration in the chamber builds up as

$$C_{\text{mes}}(t) = (C_{\text{mes}}(\infty) - C_{\text{BI}})[1 - \exp(-\gamma\lambda t)] + C_{\text{BI}}, \quad (1)$$

where $C_{\text{mes}}(t)$ is the radon concentration in the air of the measurement chamber (Bq m^{-3}); $C_{\text{mes}}(\infty)$ is the equilibrium radon concentration in the measurement chamber; C_{BI} is the radon “background” level, due to radon in air outside and to the sensitivity of the detector in the ionization chamber; λ is the radioactive decay constant of radon ($2.1 \times 10^{-6} \text{ s}^{-1}$) and γ is the relative rate of leakage given by: $\gamma = (\lambda + \lambda_v)/\lambda(-)$, with λ_v the total rate of leakage in the measurement chamber (s^{-1}). We find the equilibrium radon concentration and the relative rate of leakage by least-square fitting this equation to measured data.

For the sample geometry described in Section 2.1 and when the sample thickness is small compared with the radon diffusion length along the x -axis in the porous medium, the

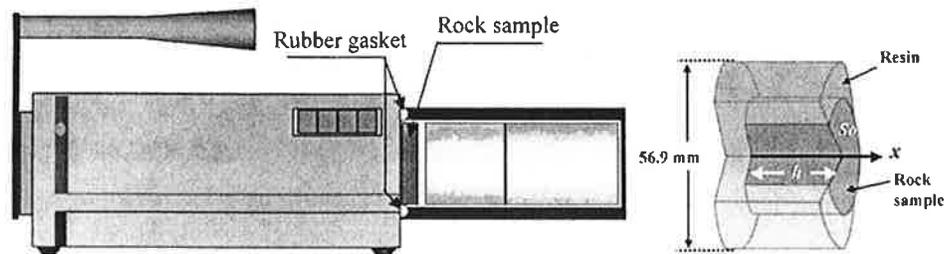


Fig. 1. The profile view of the experimental assembly with the rock sample of 0.5 cm thickness and metal cylinders of 5 and 7 cm thickness (left) and geometry of the core sample with S_0 , the free surface and h , the rock sample thickness (right).

emanation factor, $E(-)$, is found as

$$E = \frac{C_{\text{mes}}(\infty)}{\rho_d C_{\text{Ra}}} \left(p_{\text{eff}} + \gamma \frac{l_{\text{mc}}}{h} \right), \quad (2a)$$

where ρ_d is the dry bulk density of the porous medium (kg m^{-3}); C_{Ra} is the radium content of the solid material (Bq kg^{-1}); p_{eff} is the effective porosity of the medium given by (Rogers and Nielson, 1984); $p_{\text{eff}} = p(1 - s + k_{\text{H}}s) + \rho_d k_{\text{ads}}$, with p the total porosity of the medium ($-$), s its moisture saturation, k_{H} , the partition coefficient of radon in water (0.25 at 20°C) and k_{ads} , the radon sorption coefficient ($\text{m}^3 \text{kg}^{-1}$); h is the sample thickness (m) and l_{mc} is a length defined as: $l_{\text{mc}} = V_{\text{mc}}/S_0$ (m), with V_{mc} , the total void volume in the measurement chamber (m^3) and S_0 the free surface of the porous medium (m^2).

Except for strong sorbents such as activated charcoal (Schery and Whittlestone, 1989), Eq. (2a) can be reduced to

$$E \approx \gamma \frac{C_{\text{mes}}(\infty)}{\rho_d C_{\text{Ra}}} \frac{l_{\text{mc}}}{h} \quad \text{when } l_{\text{mc}} \gg h. \quad (2b)$$

Eq. (2b) implies that the term $[C_{\text{mes}}(\infty)\gamma(l_{\text{mc}}/h)]$ should be constant if evaluated for a series of experiments with the same sample material but with different experimental conditions (i.e. different leakage rates, different sample thicknesses lower than the radon diffusion length, or different volumes of the measurement chamber).

3. Radon emanation measurements

3.1. Rock samples

The core samples were collected by drilling in claystones (argillites) of Jurassic formation of the Tournemire site (Aveyron, France), where the French Institute for Nuclear Protection and Safety (IPSN) carries out research aimed at determining the confining properties of the indurated argillaceous formations (Cabrera et al., 2001). The argillite presents a very low porosity, ranging between 4% and 6% (Hg accessible porosity) and its saturation by water is close to 100%. The mean and the standard deviation of dry bulk densities and of radium contents measured in the claystone formation are, respectively, $2510 \pm 80 \text{ kg m}^{-3}$ and $33 \pm 3 \text{ Bq kg}^{-1}$. Absorption is negligible in a material close to moisture saturation (Schery and Whittlestone, 1989). The axis of the cores is parallel to the bedding plane of the claystone in order to get the highest radon diffusion length in the x -axis direction.

3.2. Preliminary tests

The mean radon “background” level (C_{Bl}), due to radon in air outside and to the sensitivity of the detector in the

ionization chamber, was based on 10 days measurements. It was $25 \pm 8 \text{ Bq m}^{-3}$ in air outside (chamber opened); also $25 \pm 8 \text{ Bq m}^{-3}$ in the air of the empty chamber and in the air of the chamber filled with metal cylinders.

The quality of the resin was verified by measuring the radon concentration increase in the cylindrical chamber after introducing in turn a block of resin (no claystone), a sample of claystone (normal configuration), a sample of claystone completely surrounded with resin (no free surface). The comparison with the background level measured in the chamber when it contained a sample of resin showed that the rock sample completely surrounded with resin made no measurable contribution to the radon concentration in the chamber. We thus verified that radon migration in the resin was negligible.

3.3. Experimental results and interpretation

The “true” value of the radon emanation factor in the rock is obtained by using expression (2b) only if the thickness of the rock sample is lower than the radon diffusion length along the x -axis in the rock. The radon concentration in the closed chamber was measured for different rock sample thicknesses and different volumes of the measurement chamber. Fig. 2 shows the radon concentration increase monitored in the measurement chamber and the theoretical curves calculated for no leakage ($\gamma = 1$) and for the best estimation of the relative rate of leakage. Long-day fluctuations of experimental data around the fitted line should be due to fluctuations of the leakage and of the radon concentration in air outside around the mean values. Table 1 reports the geometry of each experiment, the characteristics of the theoretical curve resulting from calibration to the experimental curve, and the emanation factor resulting from (2b).

Fig. 3 shows that the “apparent” radon emanation factor measured increased when the rock sample thickness decreases. This tendency is due to the loss of radon by decay during its diffusion in the rock sample and reveals that the sample thickness in experiment A and B (2 and 1 cm) is higher than the radon diffusion length in the claystone. On the contrary, the values of 0.14–0.16 carried out using a rock sample thickness of 3 and 5 mm, would represent a correct determination of the radon emanation factor in the water-saturated claystone studied.

Moreover, the inflection point of the curve in Fig. 3 leads to a radon diffusion length greater than 0.5 cm and strictly less than 1 cm, which corresponds to a radon diffusion coefficient between 5×10^{-11} and $2 \times 10^{-10} \text{ m}^2 \text{ s}^{-1}$. These values are consistent with radon diffusion coefficients observed in crystalline and metamorphic rocks reported by Tanner (1980). The diffusion coefficients of tritiated water, measured parallel to the bedding plane in these claystones, ranged between 10^{-12} and $10^{-11} \text{ m}^2 \text{ s}^{-1}$ (Cabrera et al., 2001).

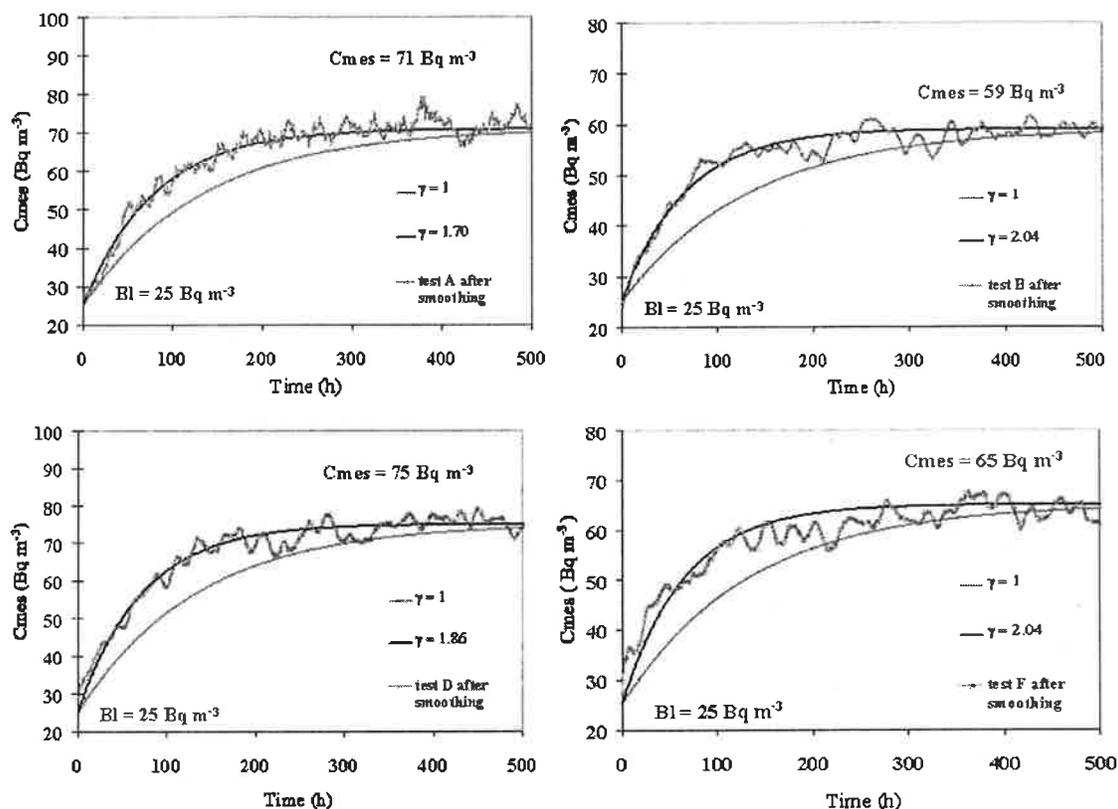


Fig. 2. Radon concentration increase measured in the chamber containing the rock sample, smoothed data and theoretical curves for each experiment. A (2 cm thick rock sample), B (1 cm thick rock sample), D (0.5 cm thick rock sample) and F (0.3 cm thick rock sample).

Table 1
Characteristics of each experiment and the “apparent” radon emanation factor measured

Experiment	h (cm)	V_{mc} (cm^3)	l_{mc} (cm)	$C_{mes}(\infty) - C_{BI}$ (Bq m^{-3})	γ	E_{mes}
A	2	900.3	94	46	1.70	0.04 (± 0.01)
B	1	922.6	96	34	2.04	0.08 (± 0.02)
C	0.5	941.7	98	28	2.46	0.16 (± 0.05)
D	0.5	624.7	65	50	1.86	0.15 (± 0.04)
E	0.5	624.7	65	40	2.52	0.16 (± 0.04)
F	0.3 ^a (0.6)	642.3	84	40	2.04	0.14 (± 0.04)

^aFor this experiment, the two faces of the rock sample were free of resin. The total thickness of the rock sample was 0.6 cm. It is equivalent to a thickness of 0.3 cm as for the comparison with the radon diffusion length in the claystone. The measurement uncertainty due to the relative rate of leakage estimation and to the radon concentration measure is indicated in brackets.

4. Conclusion

This paper describes an experimental method for determining radon emanation in undisturbed samples of consolidated porous media such as rocks or building materials. The method proposed here is based on the 21-day accumulation technique. The radon concentration is measured using an ionization chamber directly connected to the accumu-

lation container. This configuration allows the radon concentration increase to be monitored hourly without disturbance induced by air sampling. The effect of leakage on the emanation measurement is taken into account. The method was applied in order to measure the radon emanation factor in a water-saturated claystone (argillites). Four rock sample thicknesses were tested. Results showed that the value of 0.15 based on the 5 mm rock sample corresponds to

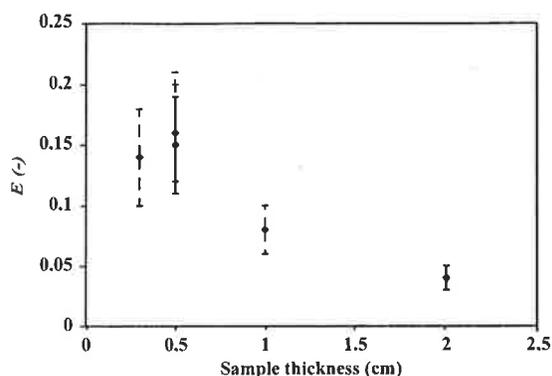


Fig. 3. The “apparent” emanation factor measured and measurement uncertainty, as a function of the rock sample thickness.

the radon emanation factor of the water-saturated claystone studied.

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